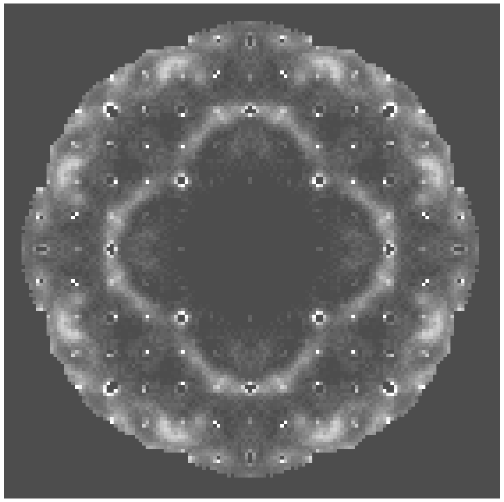
Static and Dynamic Charge and Spin Correlations in Layered Manganites

Dimitri N. Argyriou

Hahn-Meitner-Institut

Berlin, Germany

argyriou@hmi.de



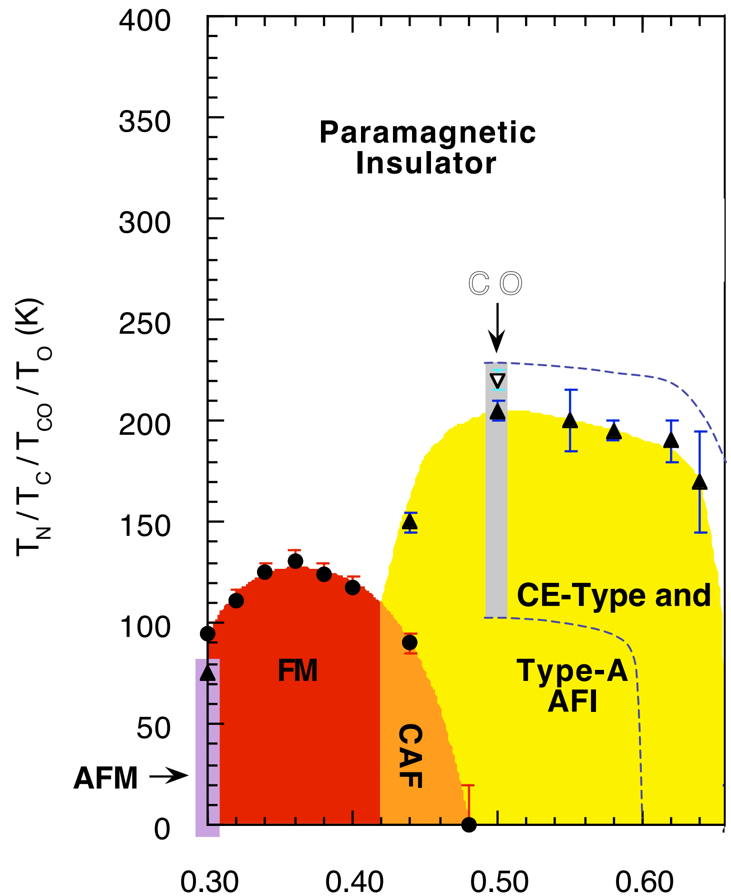
Collaborators

- ***Argonne National Laboratory***
  John Mitchell
  Uta Ruett
  Branton Campbell
  Ray Osborn
  Heloisa Nunes-Bordallo
- ***NIST***
  Jeff Lynn
- ***Hahn Meitner Institut***
  Nadir Aliouane
- ***Forshungzentrum Juelich***
  Werner Schweika

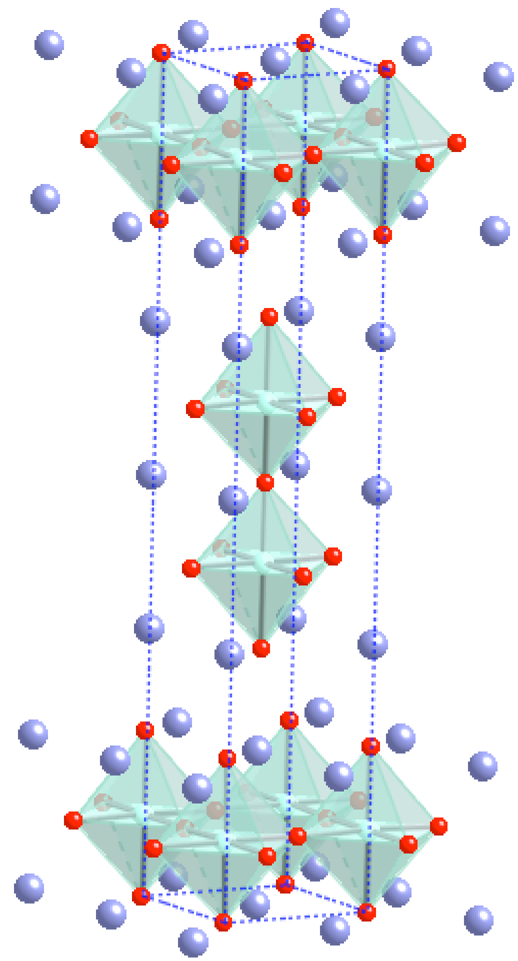
Take Home Message

- Static and dynamic charge and spin correlations are at the center of not only manganite physics but of transition metal oxide science.
  - Resurgence of manganite physics is partially due to better neutron and X-ray scattering techniques that allowed us to find evidence for lattice/spin polarons.
- Time-of-flight techniques offer unique opportunities to probe these materials.
  - Complexity requires the survey of large volumes of reciprocal space and ability to deal adeqiately with dynamical effects.

Layered Manganites $La_{2-2x}Sr_{1+x}Mn_2O_7$



x in $La_{2-2x}Sr_{1+x}Mn_2O_7$



$(A_{1-x}A'_x)_{n+1}Mn_nO_{3n+1}$

Polaron Scattering in Manganites

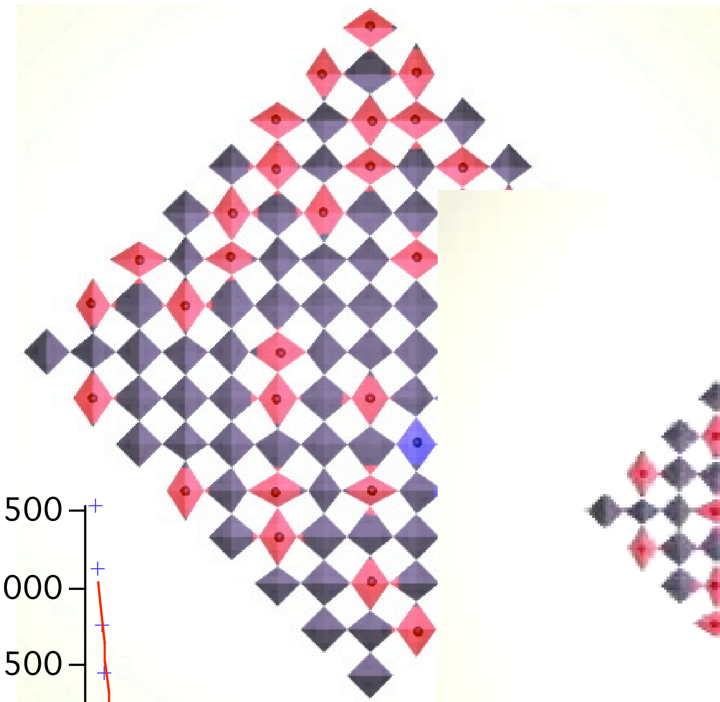
- Primary technique used is single crystal Neutron and X-ray scattering.
- Deviations from average crystal structure … ring.

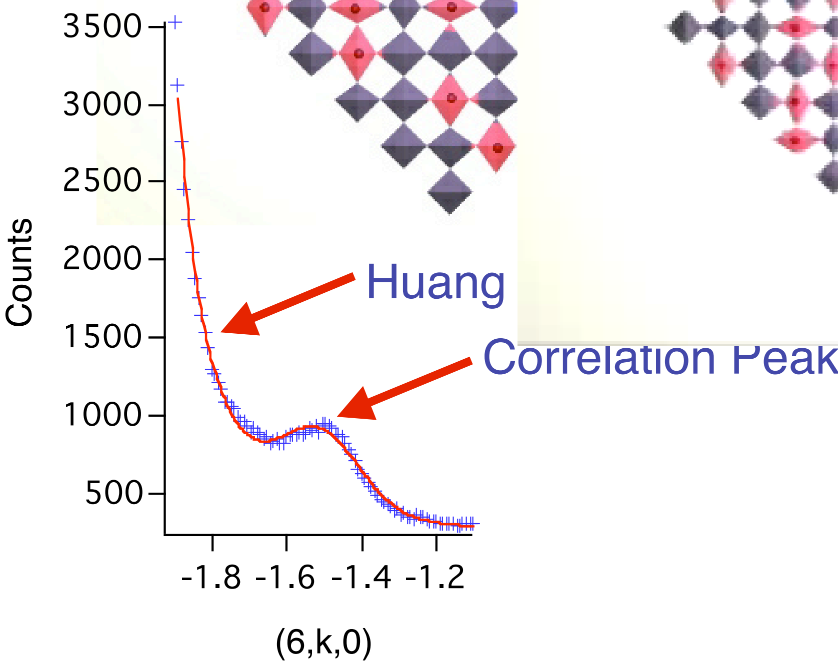
… nge lattice relaxation … s.

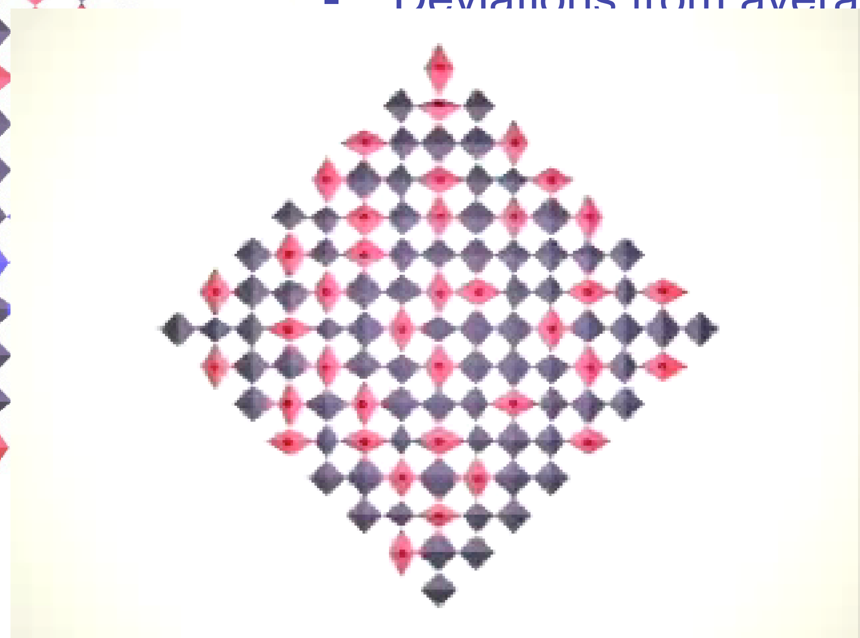
… around a Mn^{3+}

… nd Bragg reflection

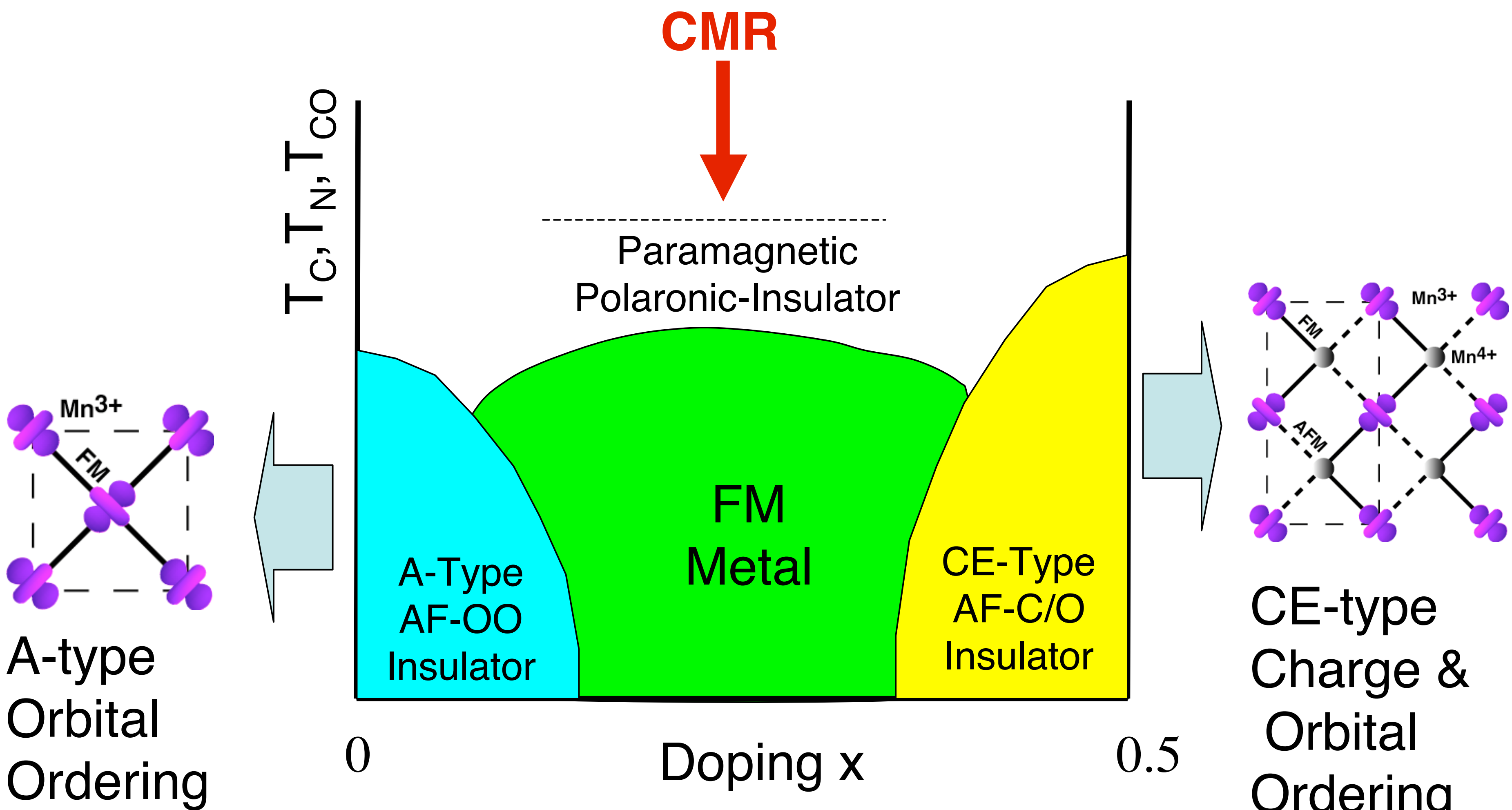
… atomic displacements … s with at particular Q-







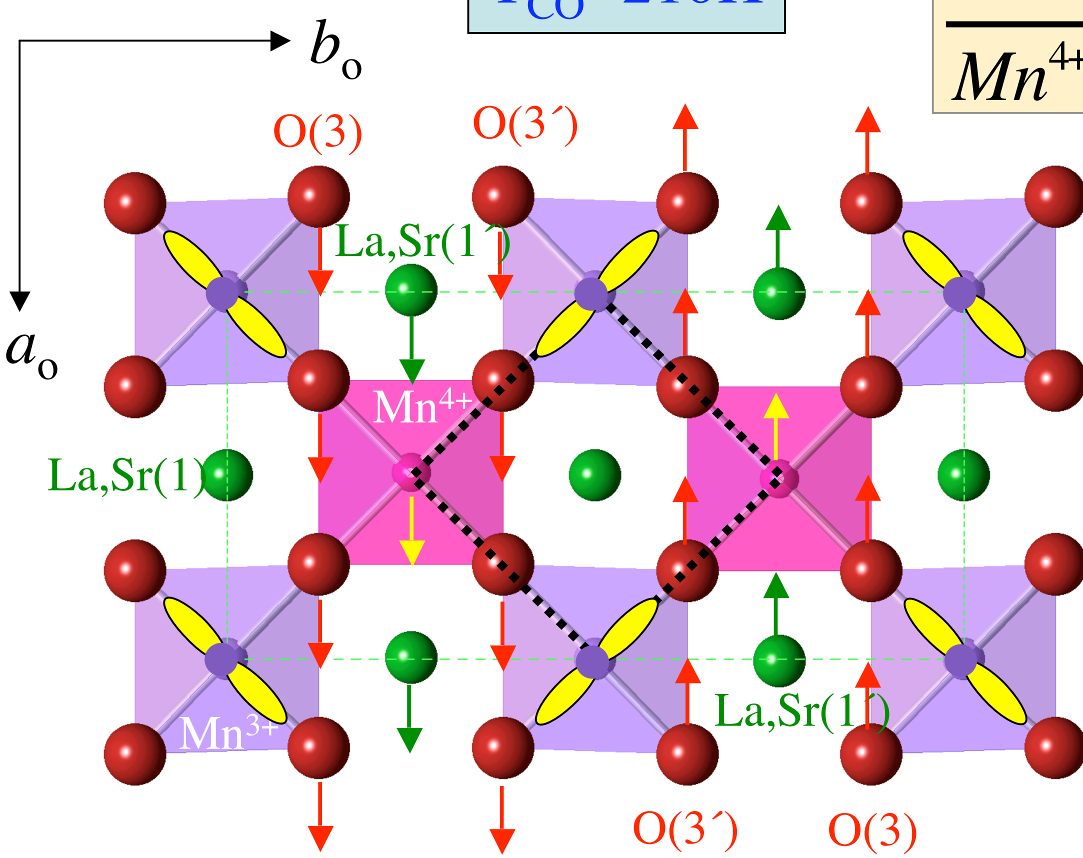
Orbital and Charge Ordering in Manganites

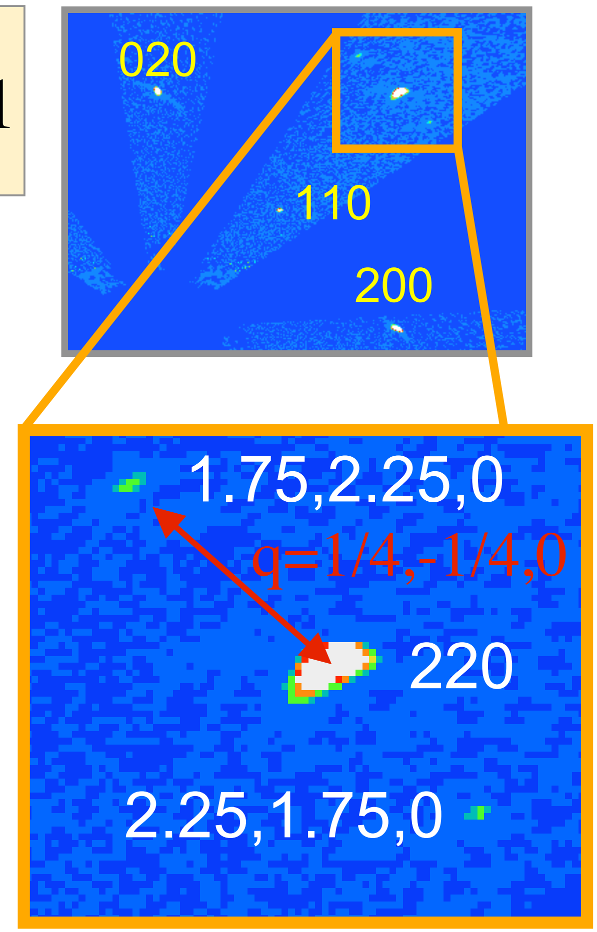


Charge and Orbital Ordering in $LaSr_2Mn_2O_7$

T_{CO}=210K

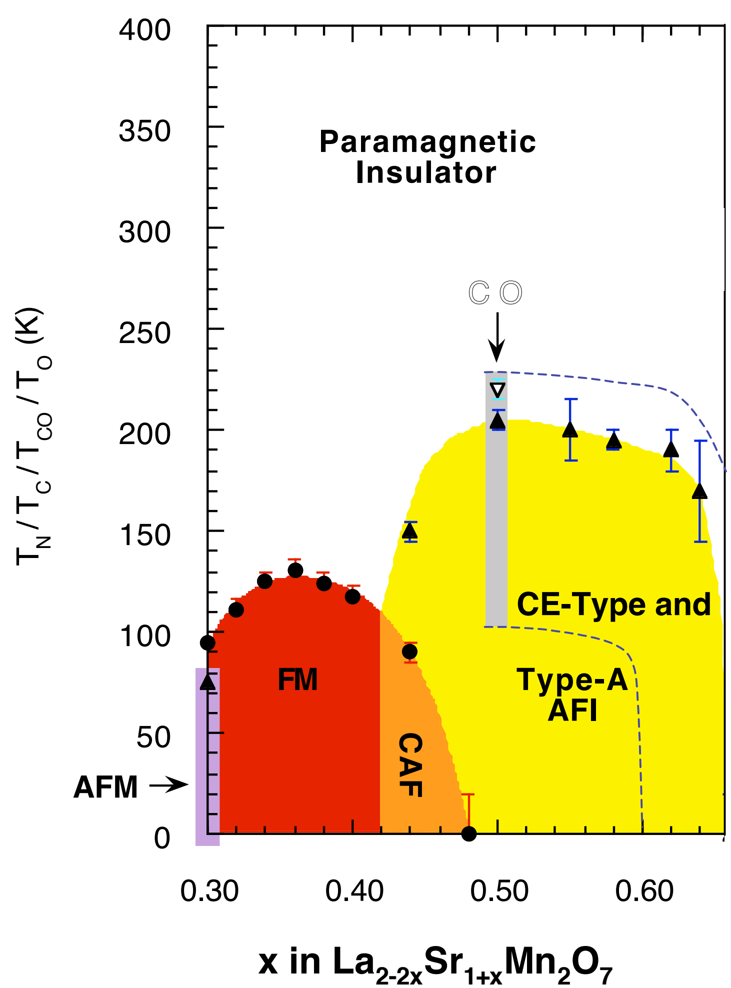
$$\frac{Mn^{3+}}{Mn^{4+}} = 1$$

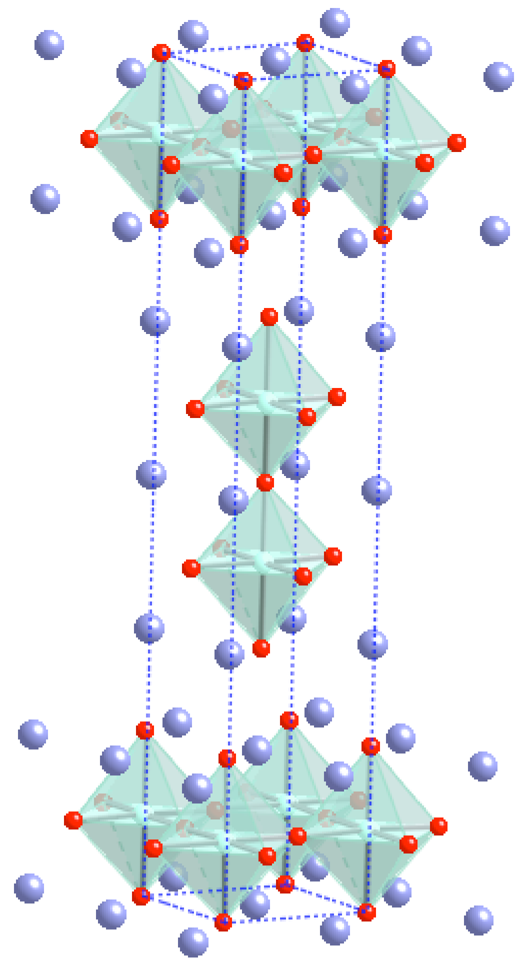




Argyriou *et al.* PRB **61**, 15269 (2000)

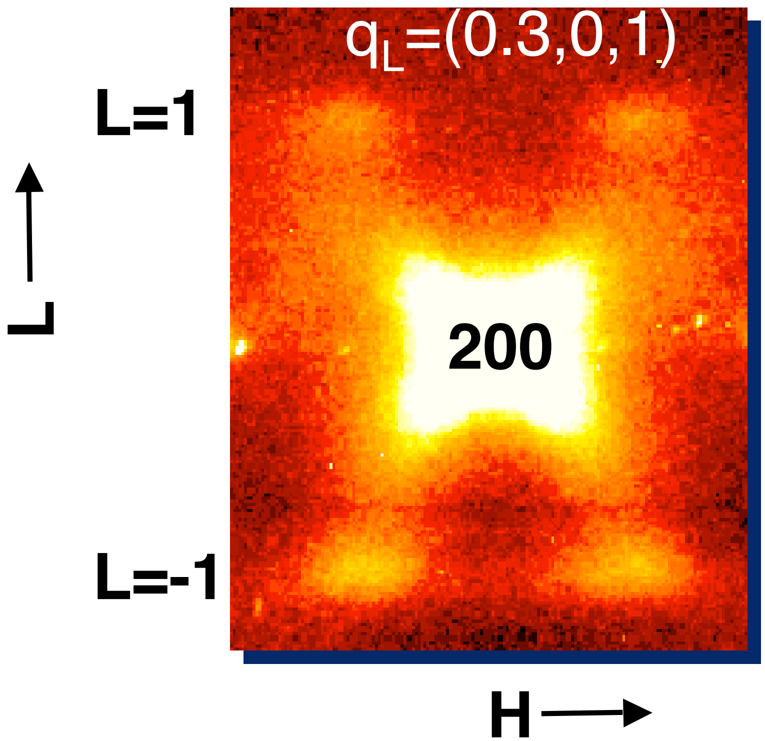
Layered Manganites $La_{2-2x}Sr_{1+x}Mn_2O_7$

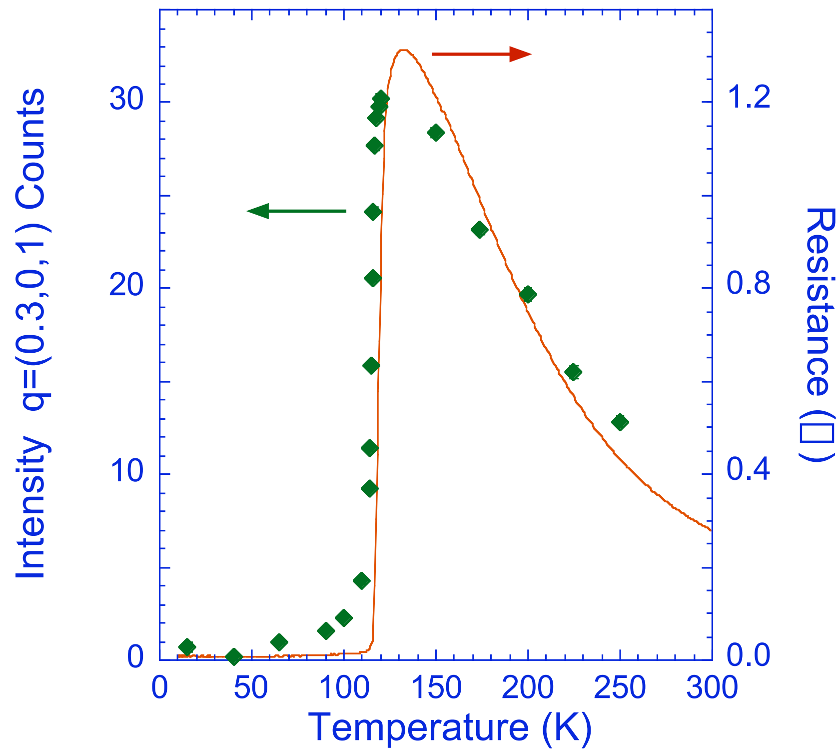


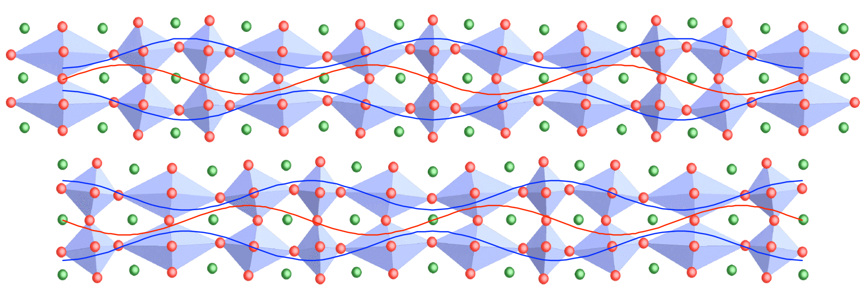


$(A_{1-x}A'_x)_{n+1}Mn_nO_{3n+1}$

Charge and Orbital Correlations in FM $La_{1.2}Sr_{1.8}Mn_2O_7$



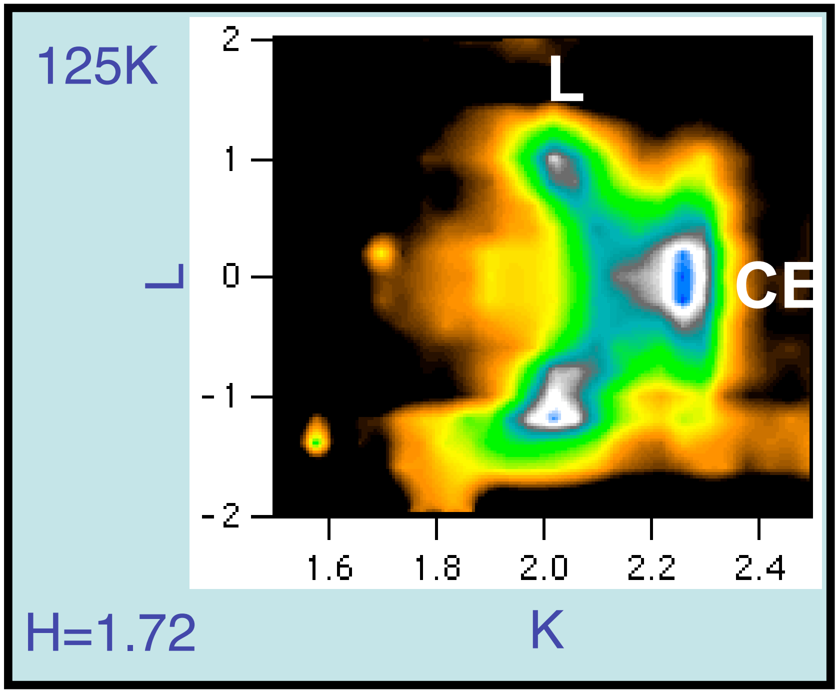


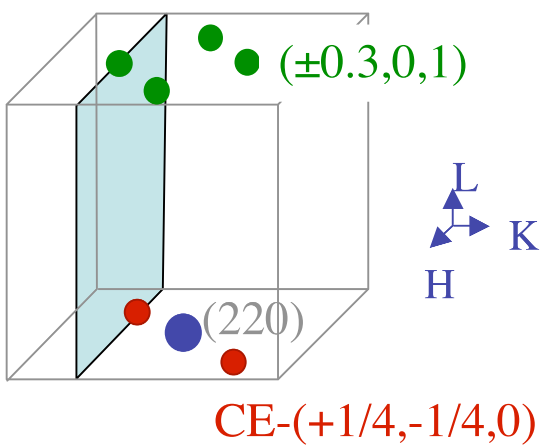


11ID-C APS
110 KV

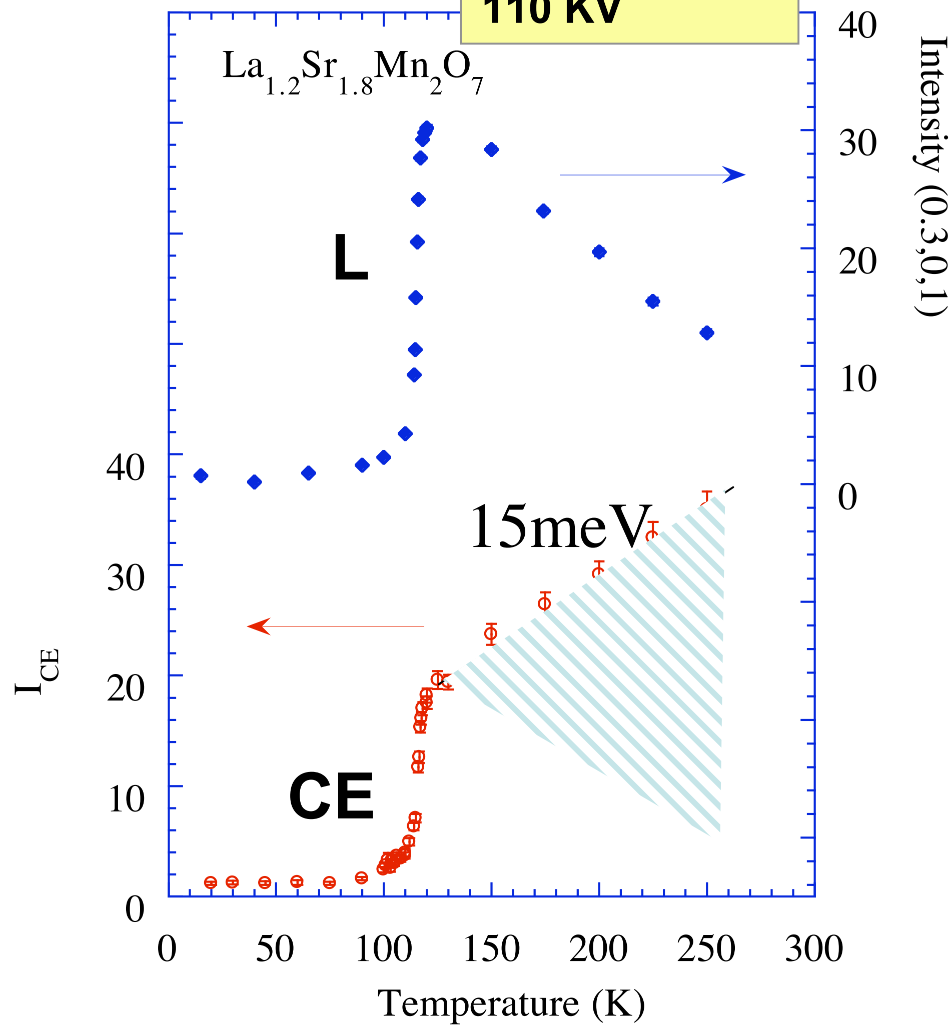
Vasiliu-Doloc *et al.* PRL, **83**,4393 (1999).
Campbell, *et al.* PRB **65**,014427 (2001).

CE-Correlations in $La_{1.2}Sr_{1.8}Mn_2O_7$ (x=0.4)

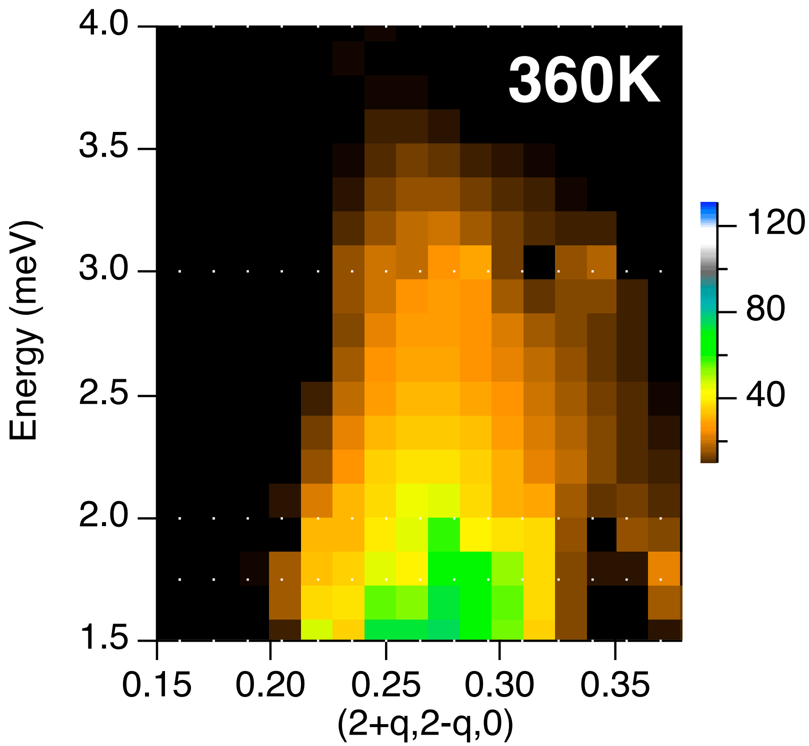


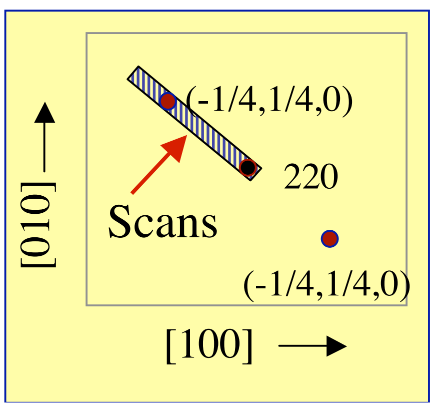


11ID-C APS
110 KV

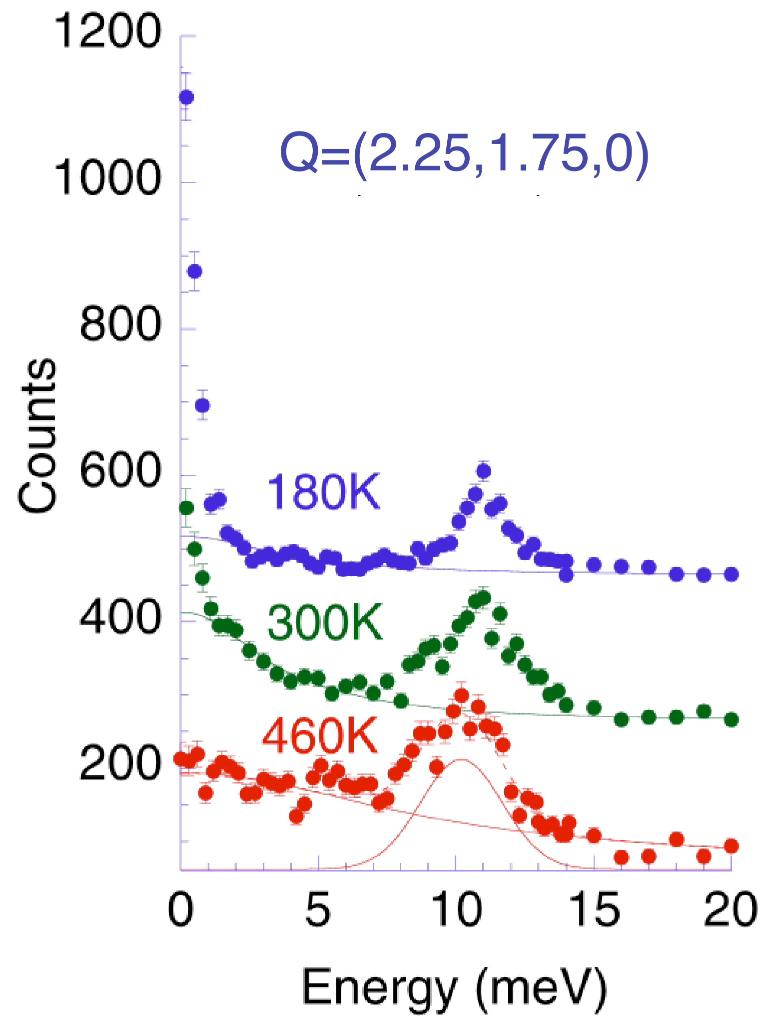


Dynamic Polaron Correlations

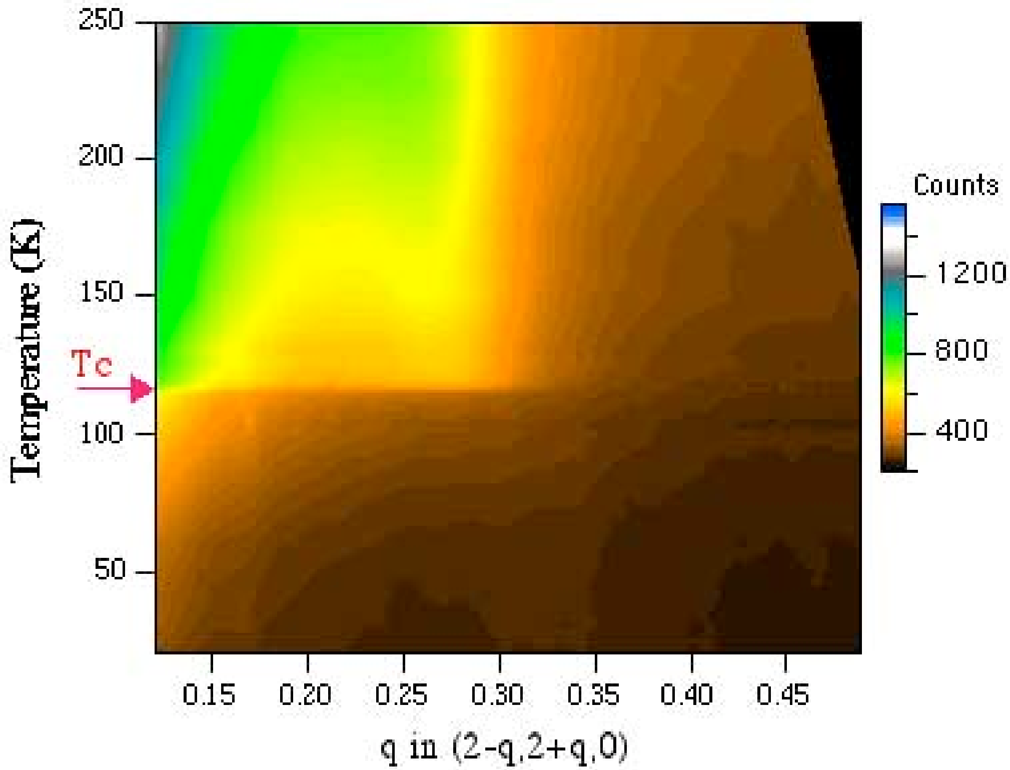


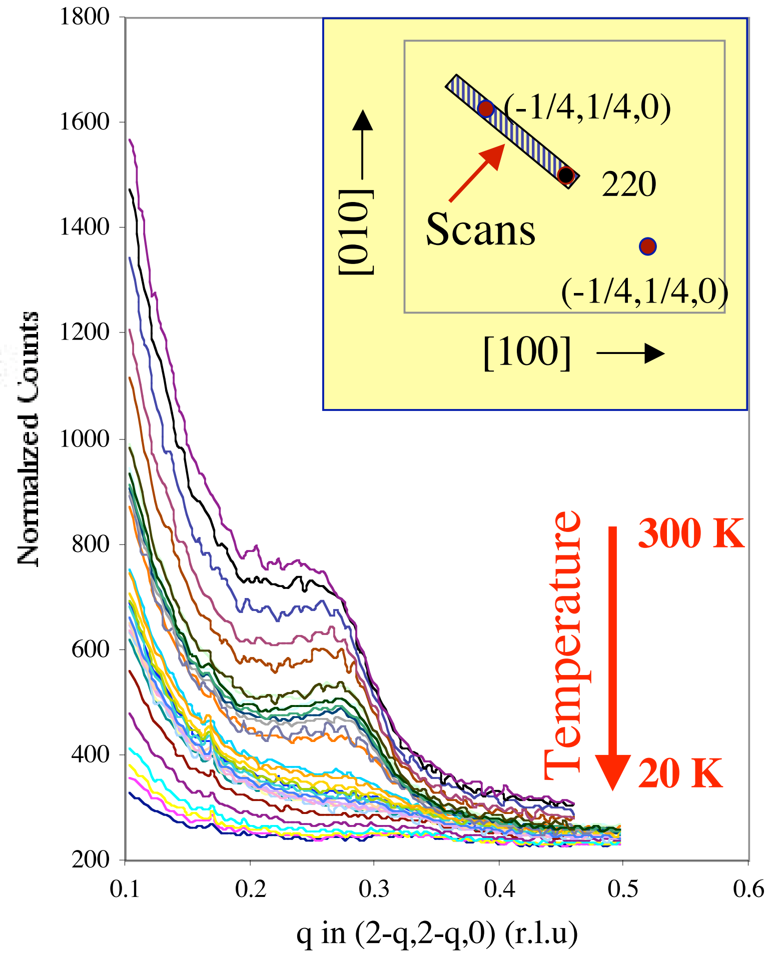


TAS, E_f=14.7meV
NIST



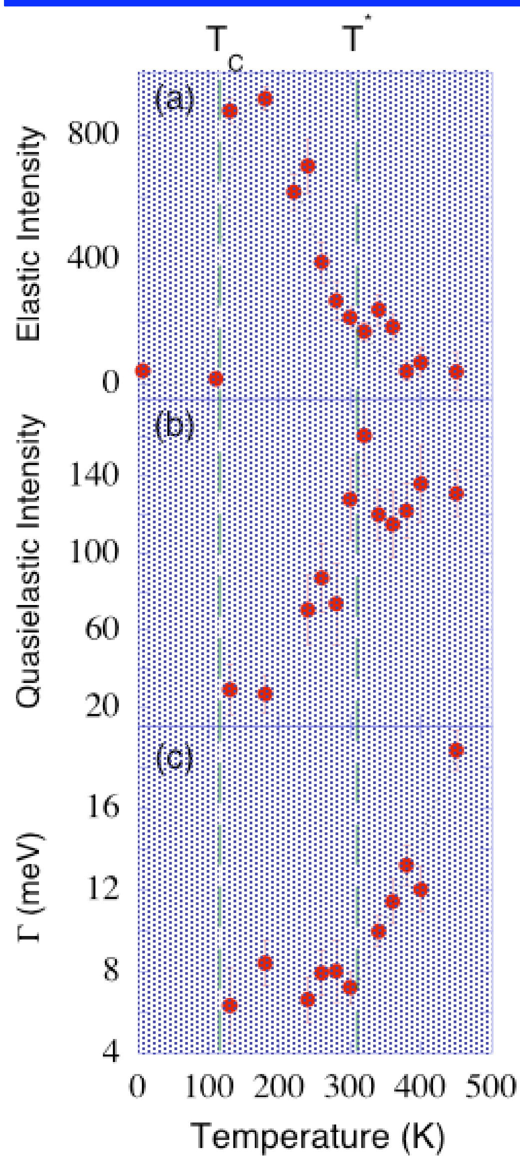
Polaron-Correlations in $La_{1.2}Sr_{1.8}Mn_2O_7$





11ID-C APS
110 KV

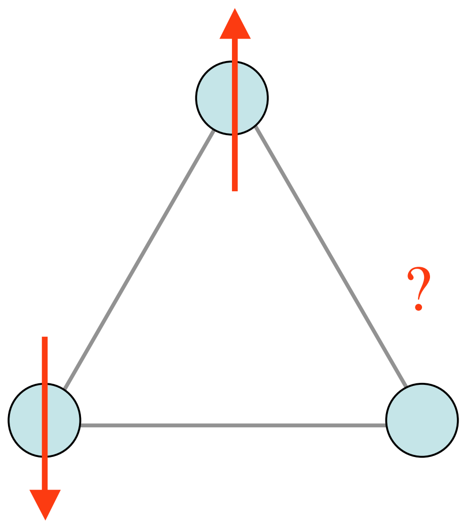
Polaronic Liquid to Glass Transition

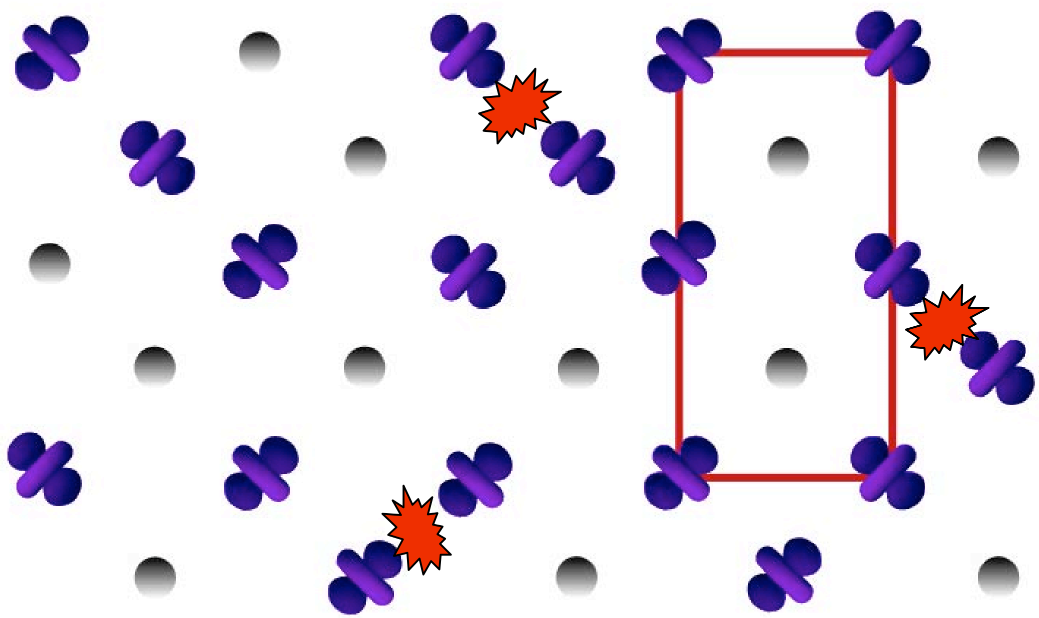


- **T>T***
  - Dynamic CE-correlations
  - Quasielastic (QE) width decreases linearly with T
- **T_C<T<T***
  - CE coherence length does not diverge
    - varies from 10-20Å
  - CE-correlations freeze
    - Elastic scattering increases below T*
    - QE scattering decreases below T*
  - QE width is ~constant (non-zero)
  - Some dynamic CE-correlations are present
- **T<T_C**
  - In metallic state elastic and dynamic polaron-correlations are not observe (no electron-phonon coupling)

Argyriou *et al.* PRL **89**,036401 (2002).

Frustration in Manganites



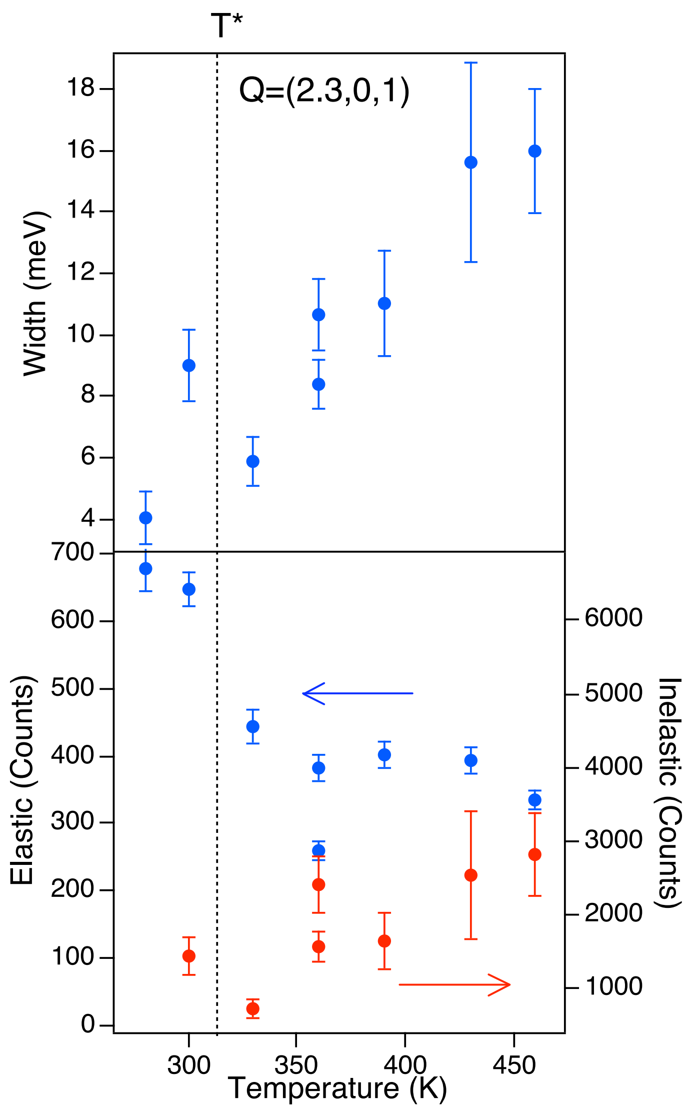


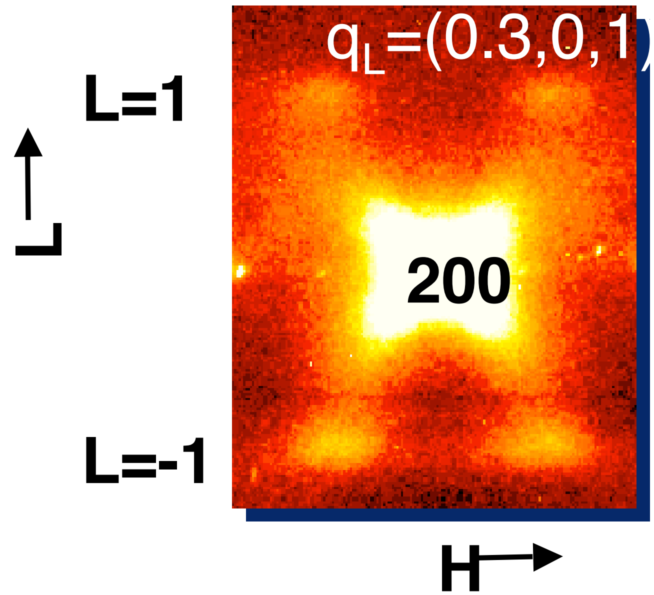
- In geometrical **frustrated** systems,
  - Critical dynamics are 2nd order-like for T>Tg.
  - Co-existence of both static and dynamic correlations.
  - At T_g non-divergent order parameter.

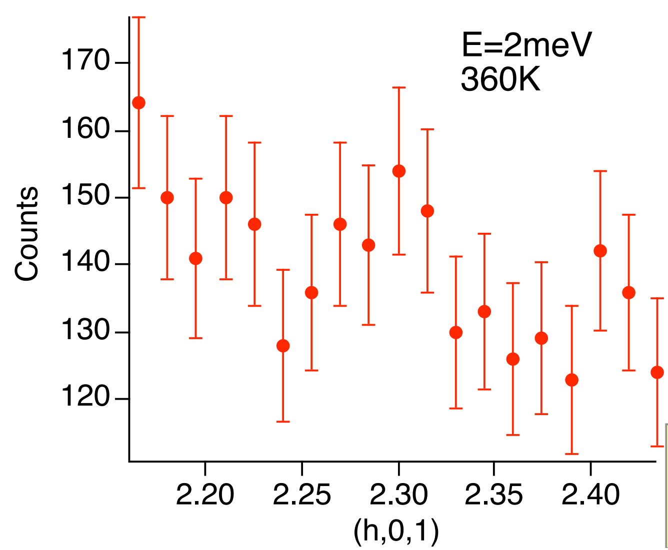
What we are actually measuring ?
Dynamic CE correlations

- Above T* *quasielastic scattering* indicates that CE-type correlations have a *finite lifetime*.
  - No evidence for diffusion of CE-type correlations.

Dynamic L-Correlations ?



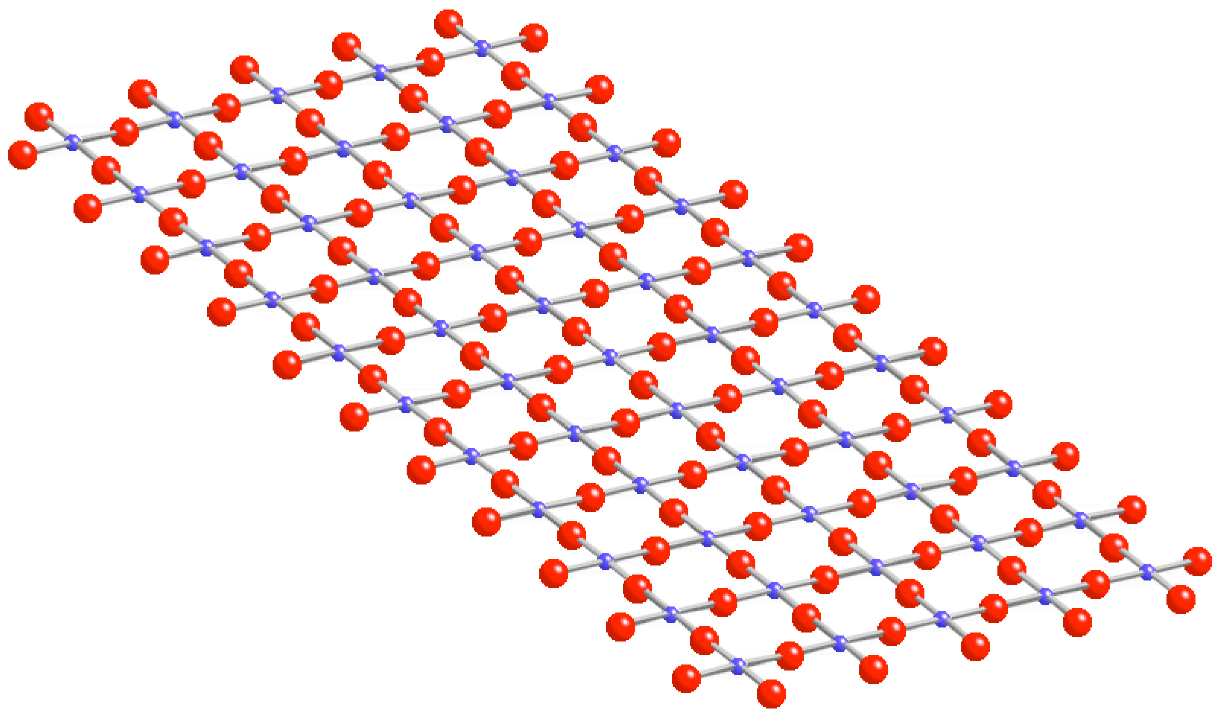


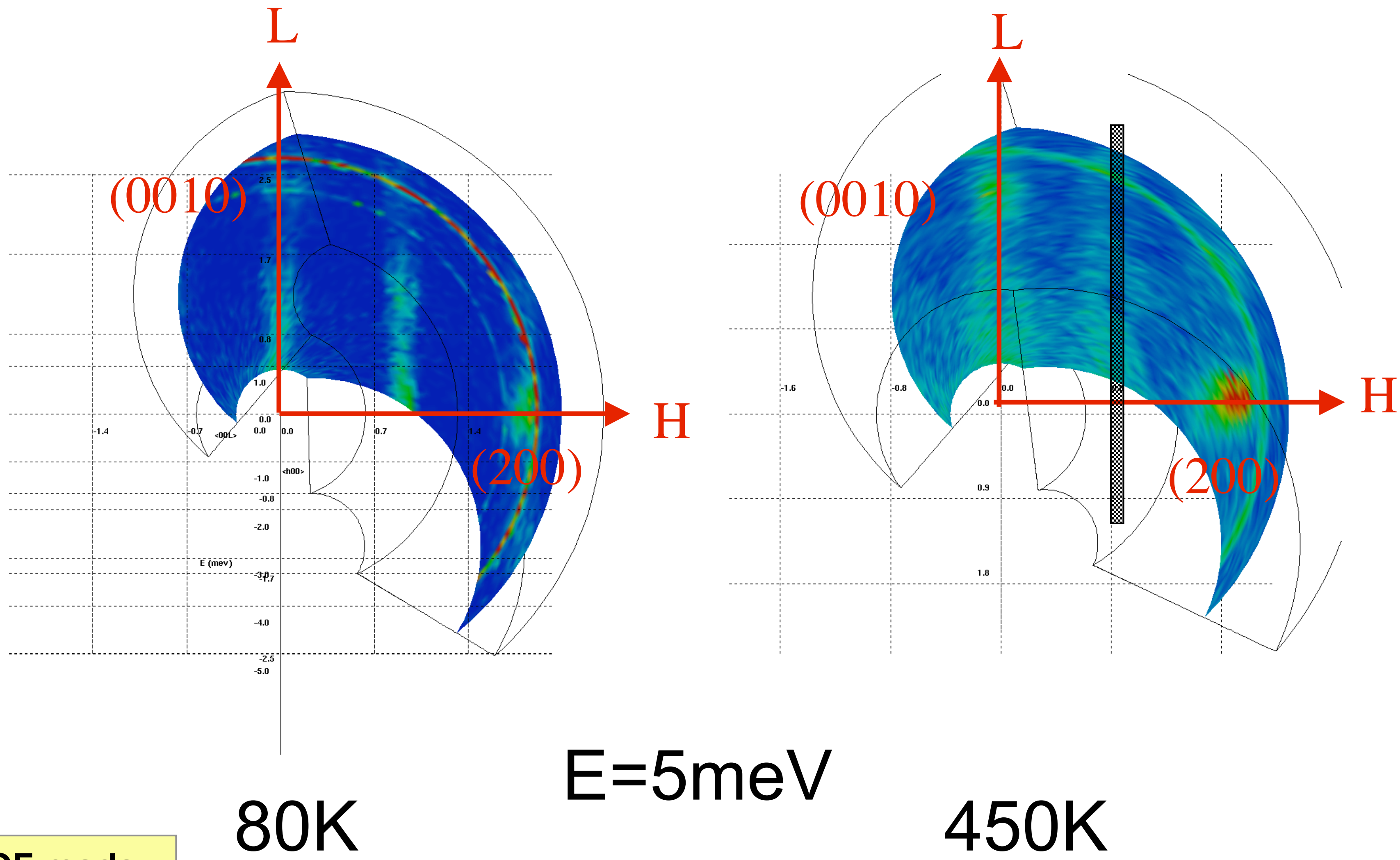


TAS, E_f=14.7meV
NIST

What we are actually measuring ?
Dynamic Lattice Relaxation

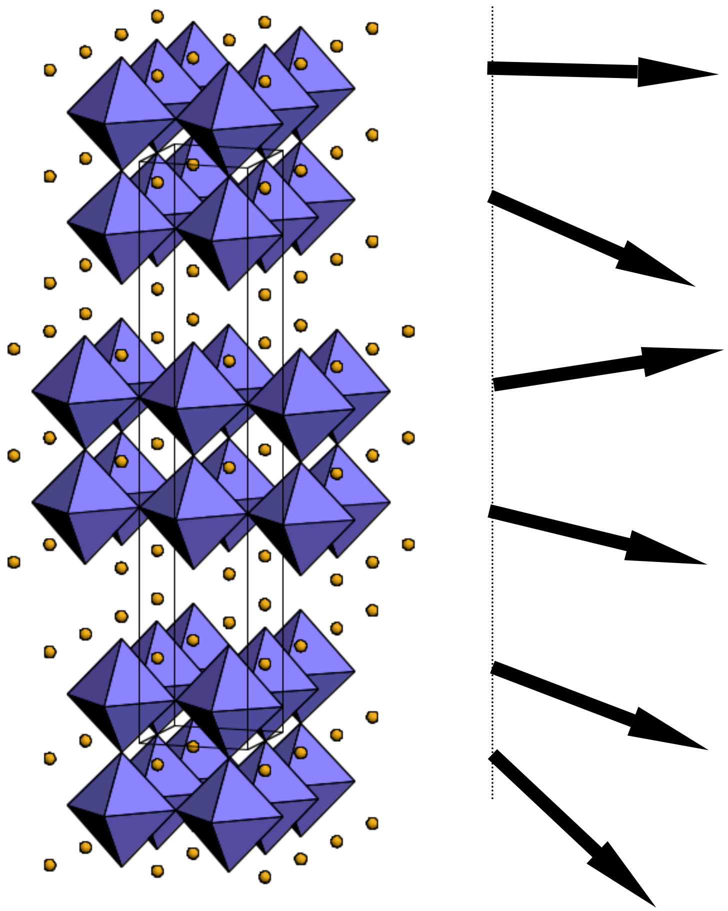
- *Quasielastic Huang scattering* indicates a dynamic lattice fluctuations associated with hoping e_g charges.

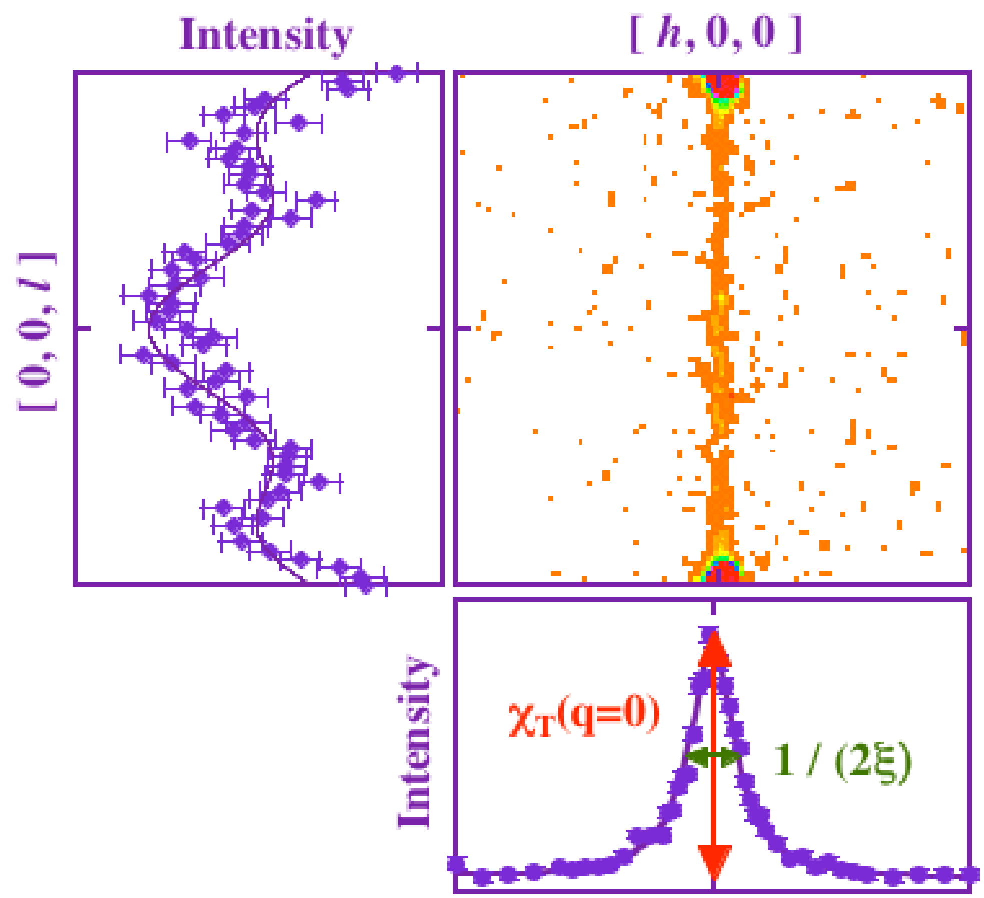




DNS, TOF mode
FZ-Jüelich

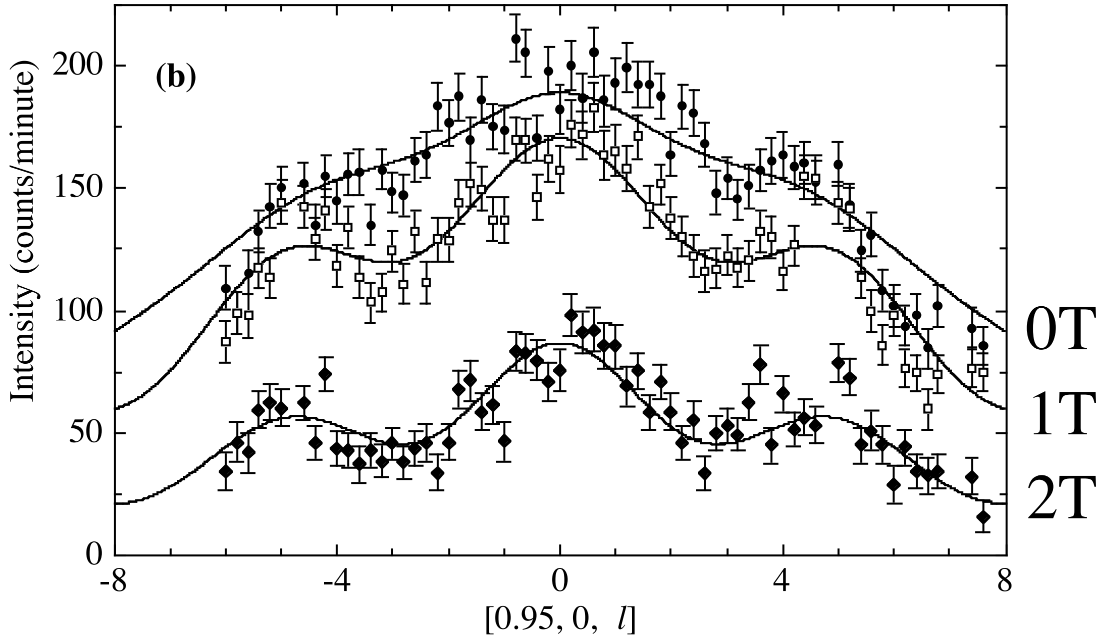
Magnetic Rods in $La_{1.2}Sr_{1.8}Mn_2O_7$





Osborn *et al.* PRL **81**,3964 (1998).

Spin Correlations in Layered Manganites

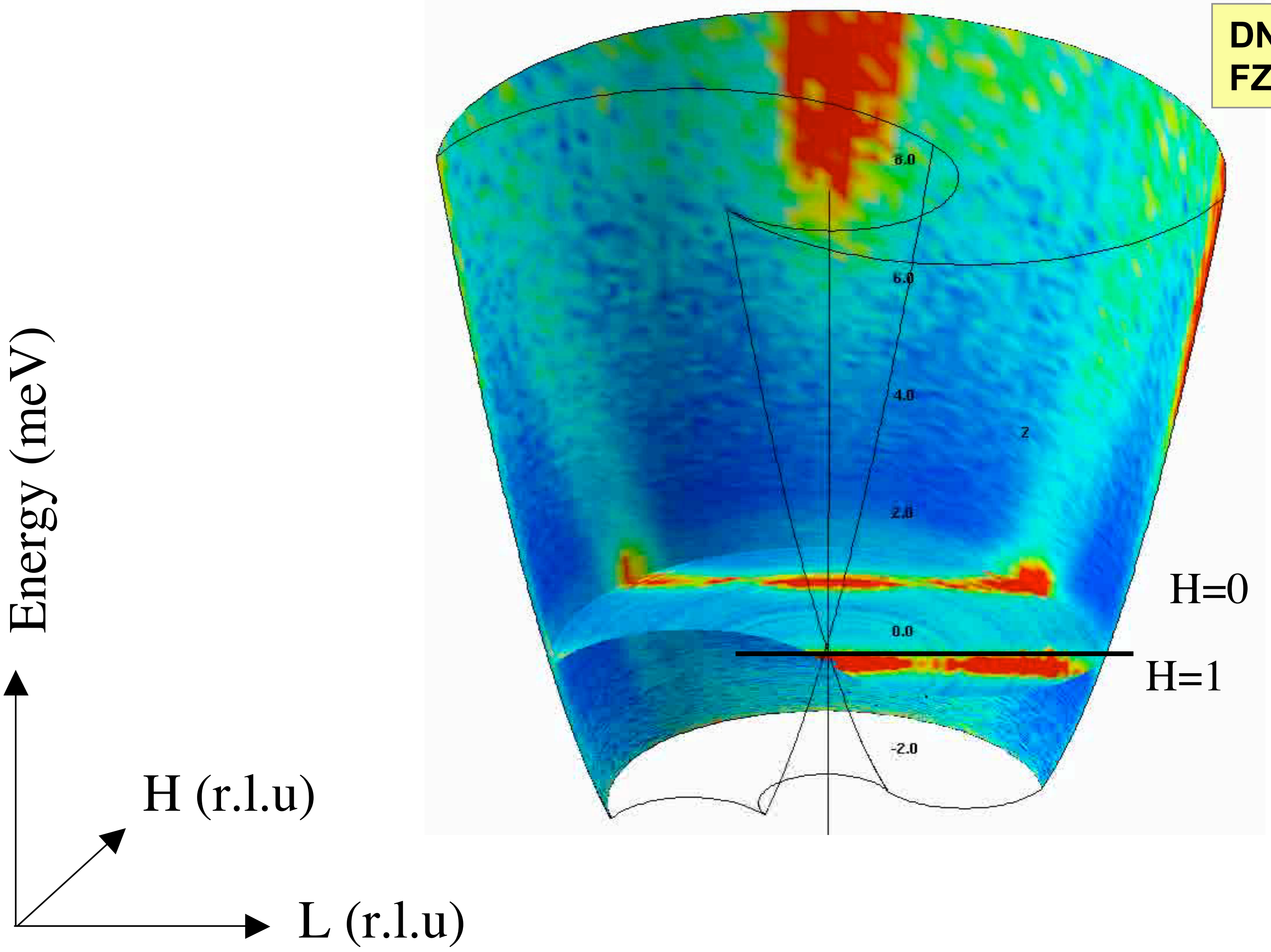


- Energy integrated scans along l show modulations in the magnetic diffuse scattering.
- The variation of intensity is modeled by spin-canting between MnO_2 sheets.
- Indicative of the competition between super- and double exchange.

Osborn *et al.* PRL **81**,3964 (1998).

Spin Correlations in Layered Manganites

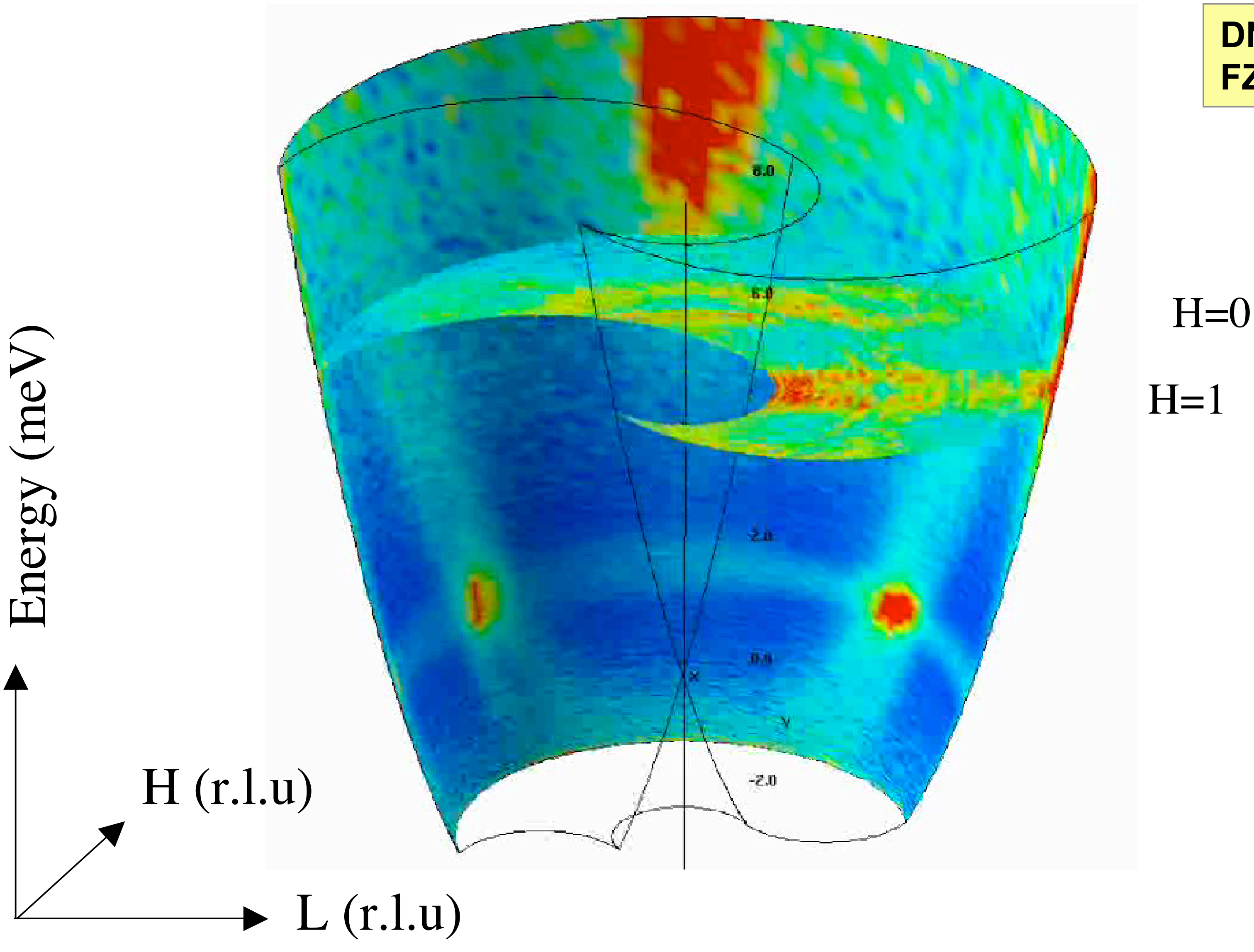
DNS, TOF mode
FZ-Jüelich



300 K

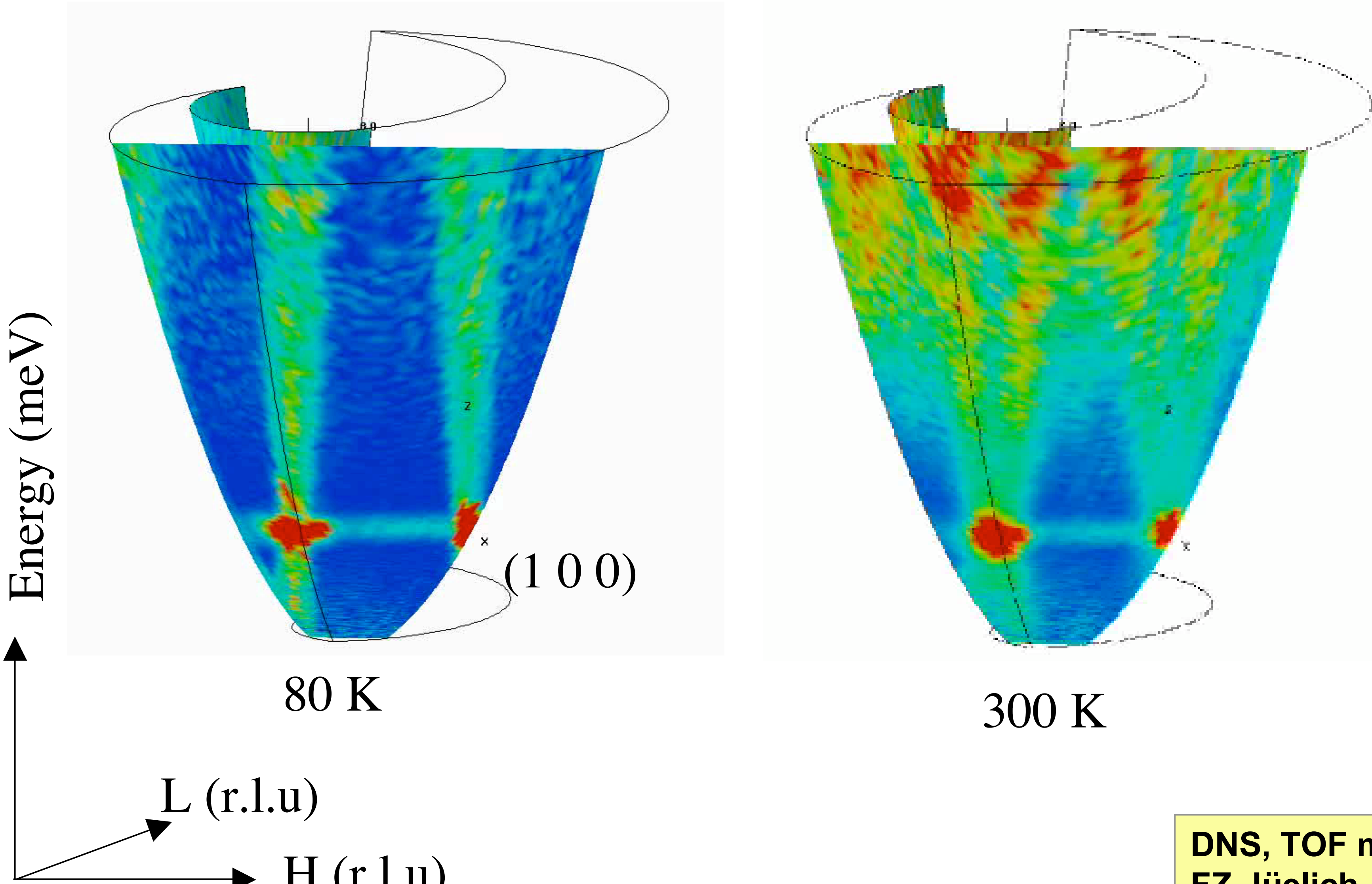
Spin Correlations in Layered Manganites

DNS, TOF mode
FZ-Jüelich



300 K

Spin Correlations in Layered Manganites



DNS, TOF mode
FZ-Jüelich

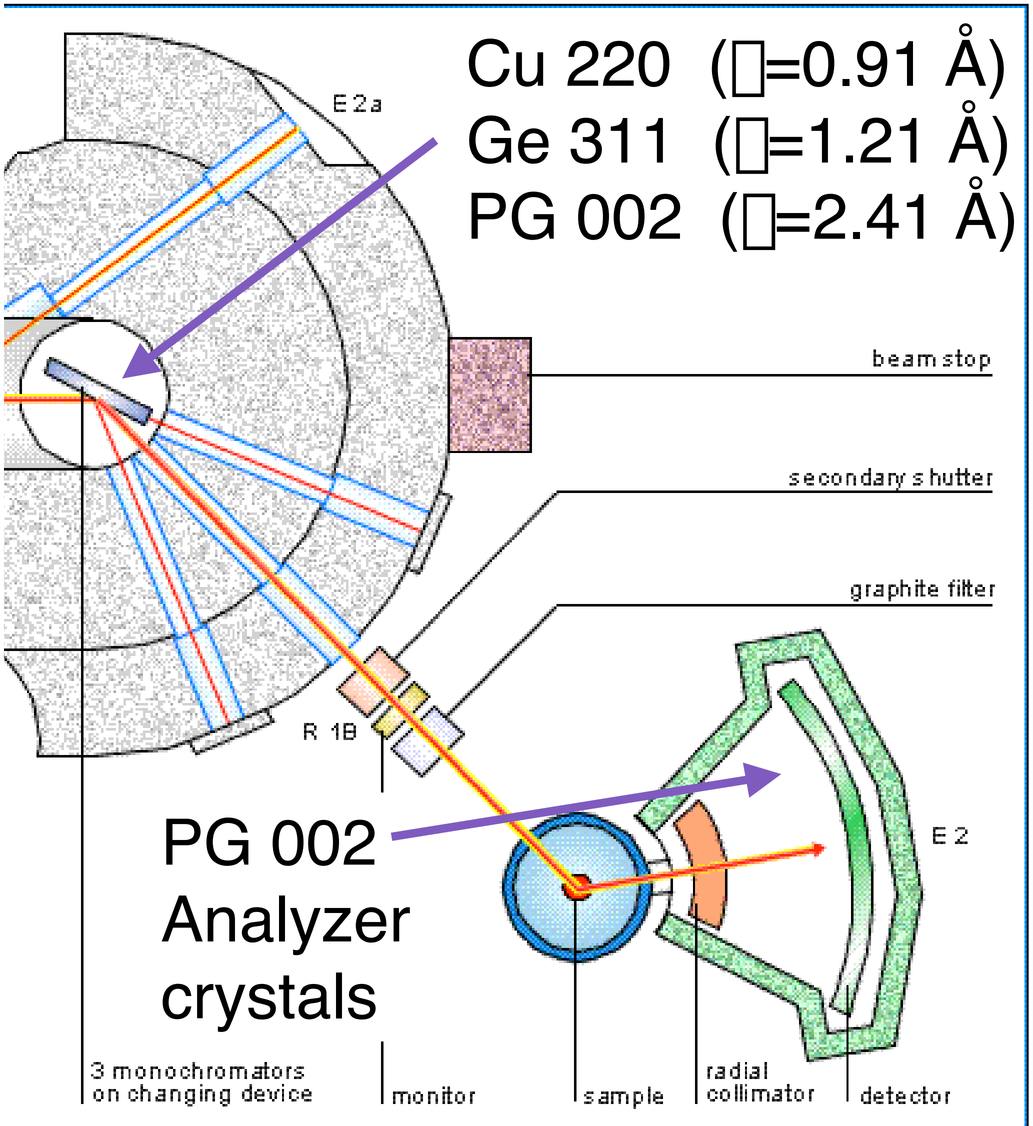
Some Lessons I

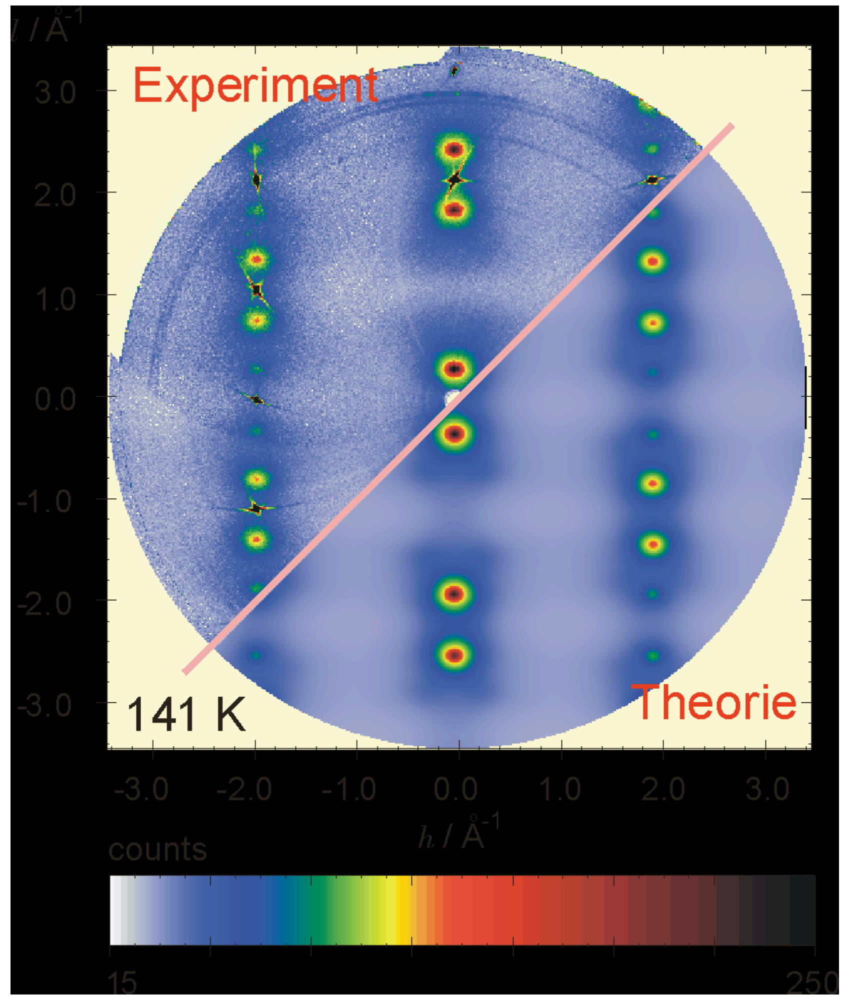
- Short range correlations are key to the physics of many materials.
  - Needs for optimized diffuse scattering instrumentation
    - Low background
    - High efficiency detectors
    - Large reciprocal space coverage
      - CCD for X-rays
      - TOF instruments for neutrons
    - Good Q resolution
      - X-rays better than neutrons
    - Energy discrimination
      - Very important background/static correlation function
      - For total scattering one needs to be aware of what is measured

Some Lessons II

- The time scale of correlations is very important and some effort should be made to probe it in an experiment.
  - X-ray scattering provides time averaged information (integration window ~1eV).
  - Energy integrated measurements provide important insights, but not the whole picture.
    - Can not see a freezing transition.
  - For neutron scattering, analyzers can be used for inelastic scattering rejection to provide the static correlation function.
    - TAS is not an efficient way to measure diffuse scattering
    - Need arrays analyzer/detectors
    - Possibilities with statistical choppers….
  - Alternatively, measure everything, sort it out later, using direct geometry time of flight techniques.

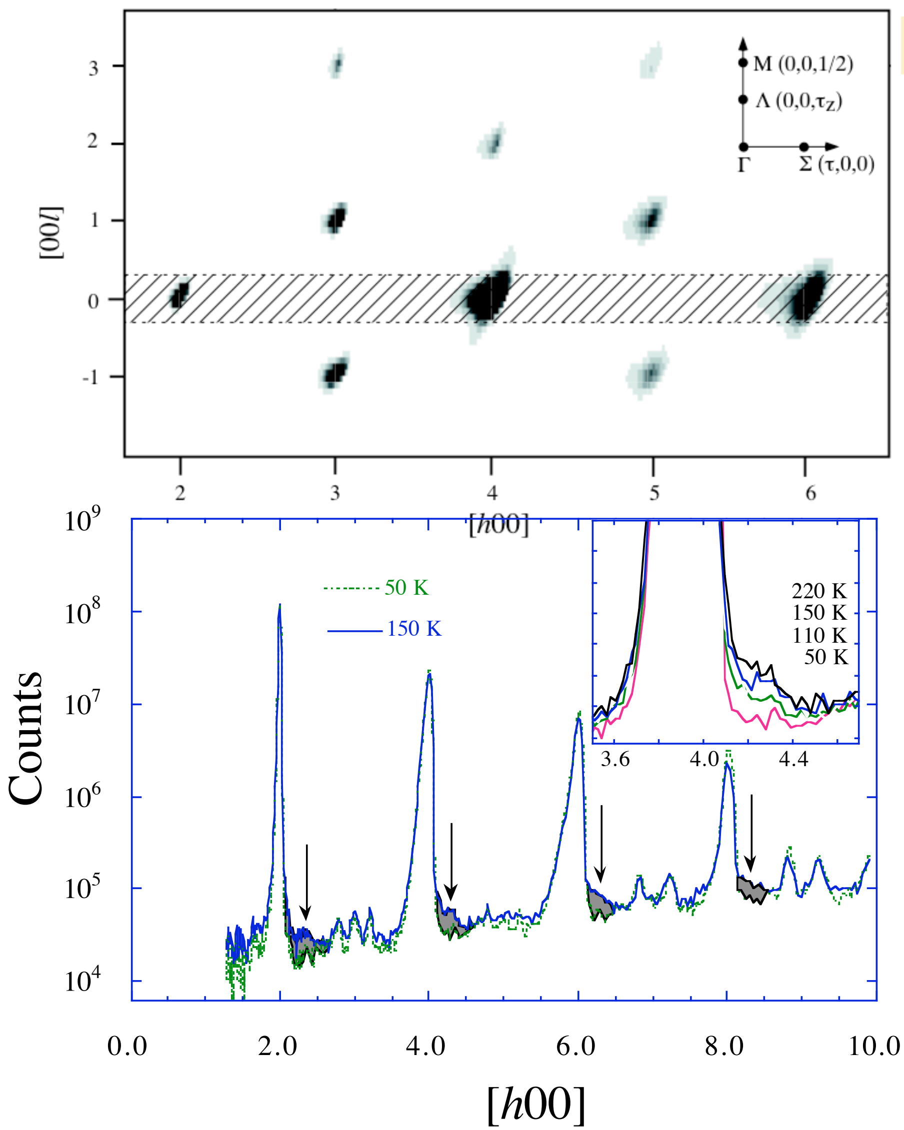
Diffuse Scattering In Rectors
E2 at Hahn-Meitner Institut

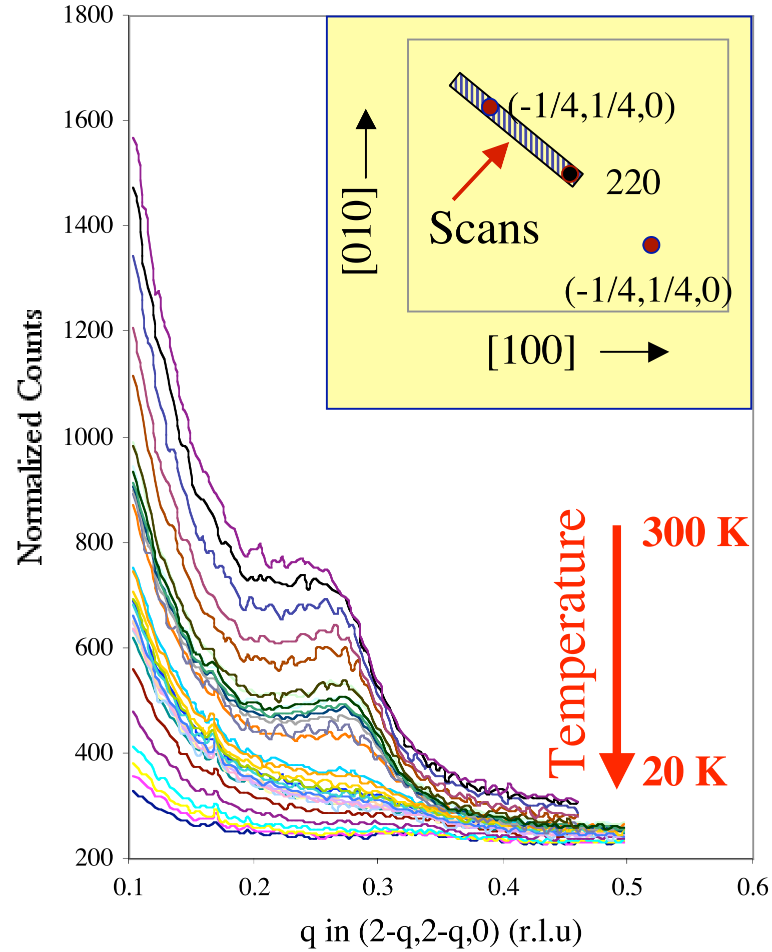




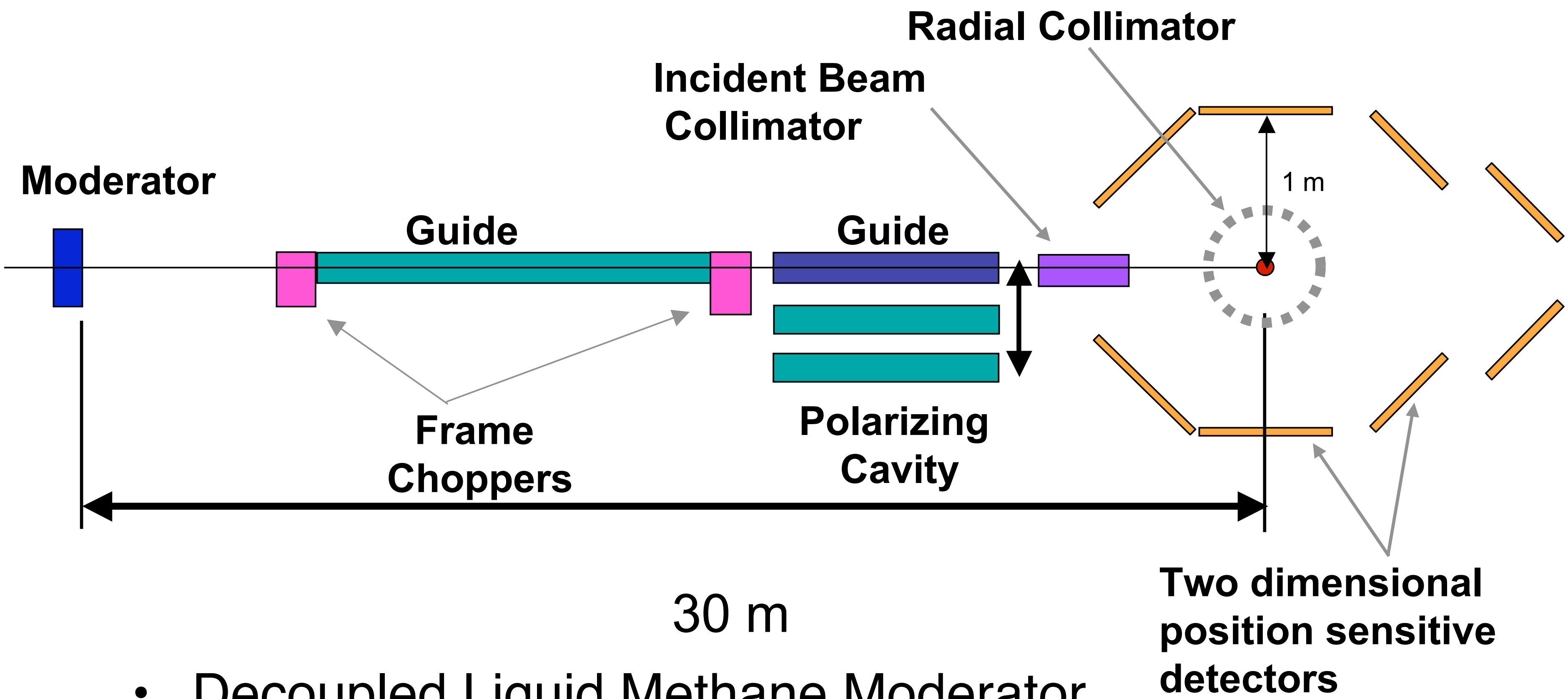
Polaron Scattering in $La_{1.2}Sr_{1.8}Mn_2O_7$ Using SCD at LANSCE

D. N. Argyriou, *et al.* , Phys. Rev. B **60**, 6200 (1999).



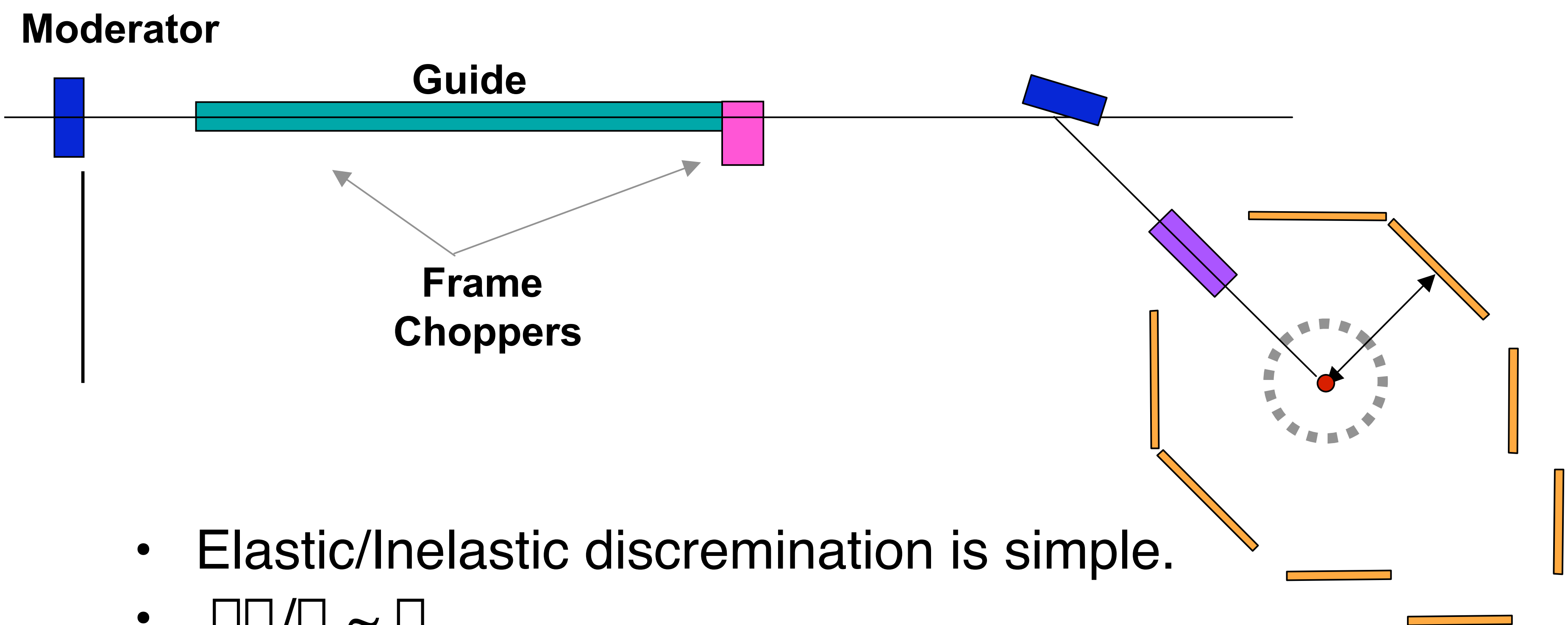


MiDaS for Long Wavelength Target Station at SNS



- Decoupled Liquid Methane Moderator
- Effective rep rate 10Hz
- Dd/d=0.2 % at backscattering.
- Single crystal and powder samples

TOF/Monochromator Combination



- Elastic/Inelastic discremination is simple.
- □□/□ ~ □
- Excellent survey instrument in Q and □ (□□=1-20 meV)
- Nuclear and magnetic diffuse scattering

Conclusions

- In $La_{1.2}Sr_{1.8}Mn_2O_7$, we find evidence of a glass transition at T*.
  - T* is the temperature were polaronic correlations freeze
  - Fundamental Temperature Scale in CMR manganites.
- The insulating state between $T_c<T<T^*$ in this manganite arises from a frozen local scale charge and orbitaly ordered state.
- Diffuse scattering has been critical in uncovering the physics of manganites.
- Quasileastic scattering gives unique insight into polaron dynamics.

Conclusions

- Instrumentation
  - X-ray and neutron diffuse scattering are complementary.
    - Should be used together whenever possible.
  - Neutrons instrumentation should focus on the better energy resolution that is achievable compared to X-rays scattering.
  - At SNS one can go beyond just measuring the static correlation function.
    - Monochromator/pulsed beam combination
    - Can be quite competitive compared to correlation shopper
    - Can bridge the gap at low energies between a diffractometer and other TOF spectrometers (like MAPS/MARI).